PHOTOINDUCED IRREVERSIBLE EFFECTS ON MAGNETIC PROPERTIES AND ALLIED PHENOMENA IN MAGNETIC OXIDES VII*

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Abstract- Photoinduced irreversible change in an optical absorption α was observed for a bulk single crystal and a thin film of yttrium-iron garnets (YIG) over the spectral range 600 or 700- 2000 nm and over the temperature range 80 -200 K. The observation is discussed based on a local structural change induced by an irradiation.

INTRODUCTION

Photoinduced magnetic effects in yttrium iron garnet (YIG) has attracted the attention of researchers since Teal et al. [1] found for the first time one of these effects, so called the I-effect. They can be divided into the I-effect (vectoral) andthe II-effect(scalar) in dependence on whether is dominant or not the polarization state of light for the observation. To the II-effect, Dillon et al. [2,3] found that at low temperatures the optical absorption coefficient α(1.06 μm) of Si^{4+}-doped yttrium-iron garnet (YIG) is decreased ($\Delta\alpha<0$) in an irreversible way by irradiation with 1.06 μm light. Here, $\Delta\alpha$ or $\alpha_a - \alpha_b$ is the difference of the optical absorption coefficient before (α_b) and after (α_a) irradiation (this effect is now called as $\Delta\alpha$-effect). They interpreted this interesting observation in terms of the different behaviors of Fe^{2+} close to and far from Si^{4+} ions. Besides,they observed that a behavior of Ca^{2+}-doped YIG showing an increase of $\alpha(\Delta\alpha>0)$ is different from that of Si-doped YIG [4]. We now confine our attention to the II-effect, following their experiment in a single crystal of a bulk and, for a comparison, a thin

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**He died on 30th January in 1994 on the tragic accident in Carpathian mountains, where he was on a ski-tour. He was born on January 16, 1960 in Kiev. In 1977 he entered Kiev university and graduated with homor in 1982. In 1987 obtained a PhD thesis entitled "Dynamics of photoinduced magnetic effects in yttrium iron garnet with Si ". In 1992 was awarded a Soros Foundation Grant from the American Physical Society, and ISF grant. Has more than 30 publications in refreed journals in which collaborated papers with us have been more than eight. He had very wide interests , besides scientific ones : sports, music , art, history and philosophy were parts of his rich internal world and every body who contacted him will remember extraordinarity of his mind. He left us , but he will remain in hearts of those who knew him.

film of YIG. Epecially, in this study, we present a more detailed discussion of the recently published study of the atomic and electronic study of photomagnetic center responsible for the II-effect[5-40]. As in our earlier study, we approach the problem of the photomagnetic center of the II-effect in two complementalry ways; phtoinduced anisotropy and photoinduced structural change. Our motivation to study the $\Delta\alpha$-effect is to clear the underlying microscoic mechanism of photoinduced magnetic effect, i.e., photoinduced disaccommodation (a gradual decrease of initial permeability with time after demagnetization) recently we observed in the similar specimens of YIG [5] .

EXPERIMENTAL

Very pure single crystals of YIG were grown by a modified floating zone method [41,42] , which have, however, a considerable oxygen vacancy V_o of the order of 0.01 mol incorporated during the process of growth, while neither Si nor Ca impurity is incorporated except for intentional dopants Ga(0.0625) . Measurement of bulk sample is difficult in the shorter wavelength region than 800 nm, since sample thickness is so large as 0.25 mm. Liquid phase epitaxial YIG films were grown on the (111) oriented $Gd_3Ga_5O_{12}$ substrate[43]. These films are transparent enough to be measured over the wavelength region 500-2000 nm , since the thickness of films is 1.0 -56.0 μm [44]. A liquid epitaxial thin film grown from PbO flax is substantially contaminated with Pb^{2+} , inducing Fe^{4+} in place of Fe^{3+}[45]. Source of irradiating the samples was a xenon flashlamp or monochromatic light. Such spectra were recorded from the longer wavelength at power levels of less than 10^{16} photons /cm^2 to avoid saturation effects. This is because the self induced effect at short wavelength is more remarkable than at the longer wavelength light. To obtain the data of $\Delta\alpha$–effect , the measurement of transmission optical density as carried out after irradiated with monochromatic or white light . To obtain the precise data of the $\Delta\alpha$–effect , the transmission optical density was recorded and simultaneously the measurement of refractive constant was carried out for the correction .

RESULTS AND DISCUSSION

Figure 1 shows the $\Delta\alpha$ at several wavelengths as a function of the cumurative numbers(n) of white-light flashes with one dose of flash $\approx 10^{19}$ photons/cm^2 at 80 K in a bulk YIG. In the case of the bulk specimens, only discreate data of transmission light may be obtained , since the penetration depth is low , *e.g.* 1 μm at 400 nm . Nevertheless, a striking change of $\Delta\alpha$ versus λ is observed as seen in this figure. Figure 2 shows the spectral dependence of $\Delta\alpha$ at 80 K, when the white light flashlights were shone 50 times on the same sample at the temperature. The maximum of $\Delta\alpha$ is seen at the measuring light of 1150 nm. A photo-induced decrease in α ($\Delta\alpha<0$) in a bulk single crystal (0.25 mm thick) is observed over 775-890 nm which is qualitatively consistent with the results in Si^{4+}-YIG[4]. It should be, however, noted that $\Delta\alpha$ over the 1000-2500 nm region is of the positive sign ($\Delta\alpha>0$) . Besides, the similar measurements in an as grown thin film of YIG (56 μm thick) were carried out at 80 K for the comparison , showing a complex behavior of $\Delta\alpha$ dependent of irradiating times. The $\Delta\alpha$-effect shows, however, the irreversibility below 200 K, or with immeasurable long relaxation time. At room temperature, the $\Delta\alpha$-effect disappears and it completely restore to the state of the samples before irradiation.

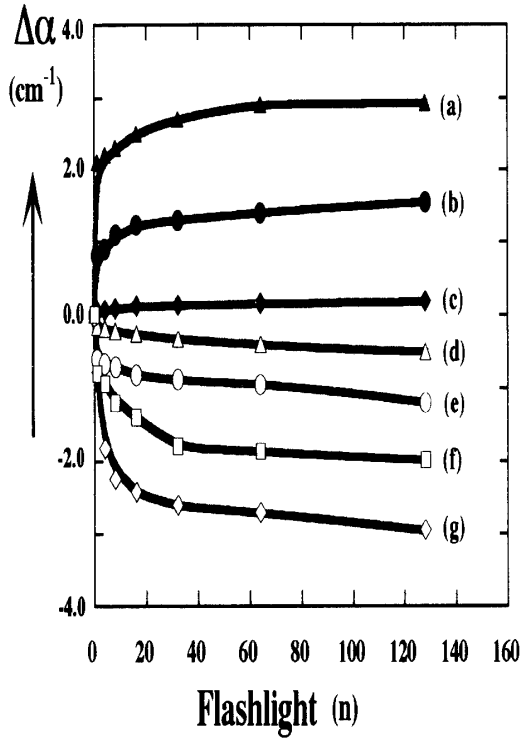

Fig.1 Δα of a bulk YIG (0.25 mm thick) at 79 K as functions of the number of the xenon - lightflashes and the measuring wavelengths. The alphabet ical letter attached with each curve means the wavelength λ of light for measuremen; (a) 1150 nm, (b) 1500 nm, (c) 950 nm, (d) 800 nm, (e) 795 nm, [f] 790 nm, [g] 775 nm.

As for the origin, we have excluded the temperature change of the specimen, since the monitored temperature was confirmed to be stable. After photoexcitation or thermal shock, there occurs a redistribution in oscillator strength, resulting in a change of optical absorption.

These correlative behavior suggests an occurrence of photoacceleration of the lattice relaxation . One of the most interesting characteristics of these phenomena is the irreversible appearance of mestastability in its properties at low temperatures,T<200 K. This metastability of properties may be a consequence of metastability of atomic configuration of garnet system inevitably involving of oxygen vacancy V_o or impurity .

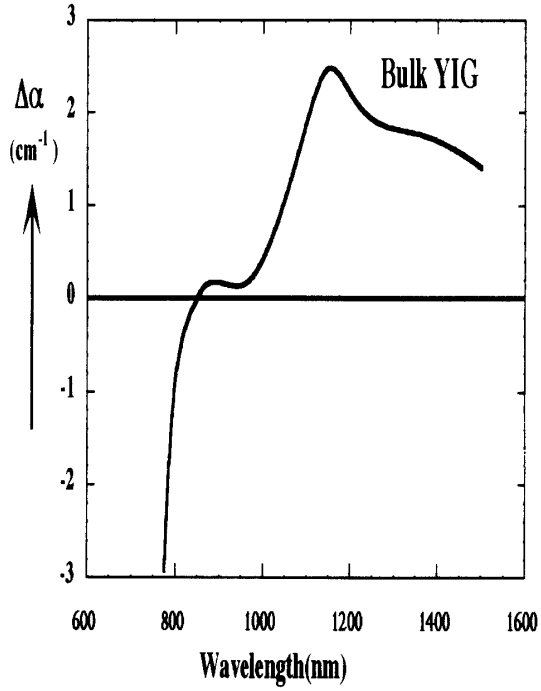

Fig.2 Spectral dependence of Δα after 50 lightflashes (full line) in a bulk specimen of YIG (0.25 mm).

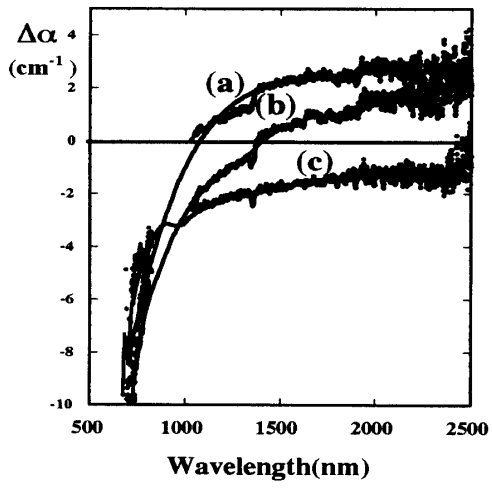

Fig.3 Δα versus wavelength for a single-crystal bulk YIG specimen at 80 K.
(a) Spectrum obtained from the 5-minute irradiation by unfocused monochromatic light (650 nm, 1 mW) (b)Spectrum obtained from the 10 minute irradiation by unfocused monochromatic light(850 nm, 0.5 mW). (c) Spectrum obatained from the difference between two measurements, just after cooling down to 80 K and 30 minutes later (withought light irradiation).

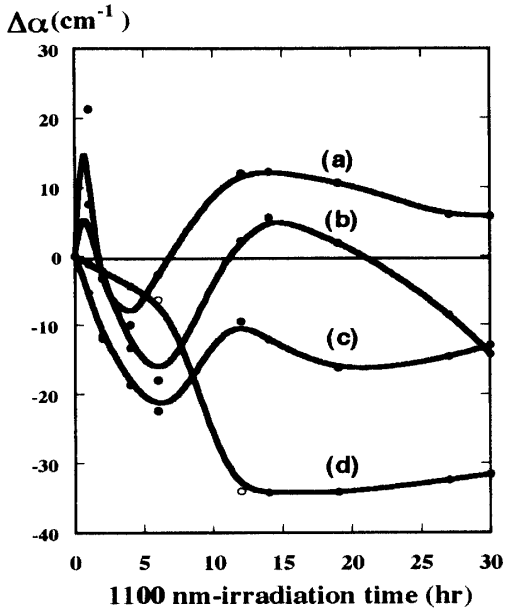

Fig.4 Irradiation time dependence of Δα at several wavelengths with exposure of monochromatic light of 1100 nm. The alphabetical letter described on each curve at which wave-length the measurement was carried out, respectively. (a) 524 nm, (b) 865 nm, (c) 750 nm, (d) 2000 nm.

A negative Δα-effect (Δα<0) in this bulk specimen is observed over 775-890 nm which is qualitatively consistent with the result of Si^{4+}-YIG [4]. It should be, however, noted that Δα over the 1000-2500 nm region is of the positive sign (Δα >0) , through the maximum and again decreases beyond 1150 nm.

The Δα–effect shows the irreversibility below 200 K, or with immeasurable long relaxation time. At room temperature, the Δα–effect disappears and it completely restore to the state of the specimens before irradiation. The fact that the critical temperature for the observation of a variety of photoinduced magnetic effects in YIG is 200 K favors the idea that the unique origin is common to these phenomena [4, 5]. Next, the similar measurements in as grown thin film of YIG (56 μm thick) were carried out at 79 K for the comparison , showing a complex behavior of Δα dependent of irradiating times and wavelength. Figure 4 is an example of these measurements under 1100 nm-irradiation. These data were corrected by eliminating the background or thermal change of Δα. In order to explain these observations , we propose a configuration coordinate model, which assumes the formation of microscopic potential barriers around oxygen vacancies, triggered by photoexcitation of electrons trapped at the sites. The irreversible characteristics may be explained by the assumption that these microscopic barriers prevent the photoexcited electrons from being recaptured, the probable origin of which is the relaxation of the crystal lattice around the vacancy [11]. It should be noted that the Δα–effect at short wavelength region , i.e., less than 1000 nm is observed also with monochromatic 1100 nm-irradiation . This result suggests that not only excited electrons but also lattice vibration change the distribution of oscillator strength in an irreversible way so long as kept in low temperatures.

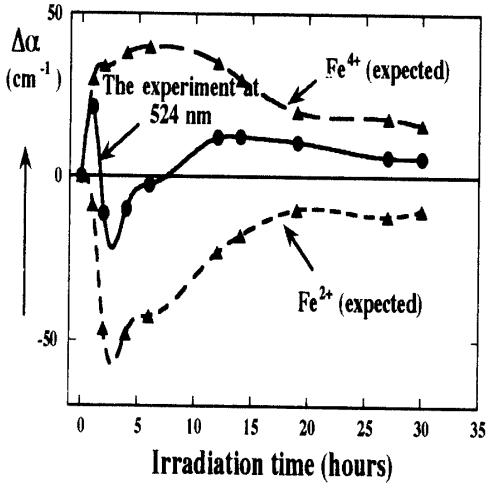

Fig.5 Superposition of both photoinduced contributions of Fe^{4+}(Δα>0) and Fe^{2+}(Δα<0) in the thin film.

As shown in Fig.5, the superposition of both competing contributions of Fe^{4+} ($\Delta\alpha>0$) and Fe^{2+} ($\Delta\alpha<0$), each of which concentrations will change in a certain range with irradiation time, could form the complicated curves, depending on the wavelengths as this is the case in Fig.4. At low temperature, oxygen vacancies V_o behave as a type of charge reservoir which consists of two electrons trapped at the sites (V_o^{2e}) or (Fe^{2+} -V_o- Fe^{2+}) and light sensitive injector of free electrons into this insulating system. That is, the photoexcitation is the alternative method of variation of the free carrier concentration. This process may be illustrated schematically in Fig. 6. In this figure, the point of A on the lowest curve corresponds to the total energy in its ground state or (Fe^{2+} -V_o- Fe^{2+}) ; the shifted concave B is (Fe^{2+} -V_o) + Fe^{2+} (nearly free) and the central curve B* is (V_o)+ 2 Fe^{2+} (nearly free) and the upper curve is (V_o) +2Fe^{2+} (free). The direct process begins with electronic excitation by a photon absorbed at V_o^{2e} situated at Q_0 (A→A*) and then through several paths at Q_1(B→B*). The process of the former (A→A*) needs the much more photon energy than that of the latter (B-→B*). Then we can explain the reason why the sign of $\Delta\alpha$ is negative at short wavelength region, although one question($\Delta\alpha/\Delta\lambda$ <0 over 1150 nm) is left. So far the discussion is limited to the situation assumed only for electrons or Fe^{2+} but may be extended for holes or Fe^{4+} A microscopic and quantitative description is quite complicated due to the interference of various interactions among electrons, holes, vacancies and impurities. The microscopic explanation must await detailed analysis of these interactions. Largely overlooked, however, in earlier analysis, we believe the study of these relaxations is helpful for understanding the photomagnetic center in YIG. The relative positions of the configuration coordinate parabolas is a crucial parameter, leading to either normal or metastable states. Such states are called large lattice relaxation states and are believed to be responsible for a broad class of metastable phenomena observed in many doped semiconductors. A typical example is the persistent photoconductivity [44]. The irreversible characteristics may be explained by the assumption that these microscopic barriers prevent the photoexcited electrons from being recaptured, the probable origin of which is the relaxation of the crystal lattice around the vacancy [45]. At low temperature, oxygen vacancies V_o behave as a type of charge reservoir which consists of two electrons trapped at the sites and light sensitive injector of free electrons into this insulating system. The equilibrim positions of the surrounding atoms will depend in general on the electronic state of the impurity , since a change in charge distribution changes the interactions with the neighbors. Thus the minima if the various energy surfaces occur at different values of the configuration coordinates. In a qualitative way we may expect the direction of the shift in ionic materials: if the impurity or vacancy is positive, the transition to an excited state extends it electronic charge distribution further away from its original position. Thus the positive ion is effectively made more positive and has a greater attraction for the nearest (negative) ions. Hence we would expect the minimum in the excited state to occur at smaller inter-ionic distances than for the equilibrium values in the ground state. When the impurity is negative ion or an oxygen vacancy trapped with two electrons , the arguments are reversed. It seems likely that, in general the curves for the excited state have less curvature near the minima than the ground state because of their more diffuse charge distributions. On the other hand, the irreversible variation in $\Delta\alpha$ is destroyed at room temperature, which is caused by the returning of electrons surmounting the potential barrier

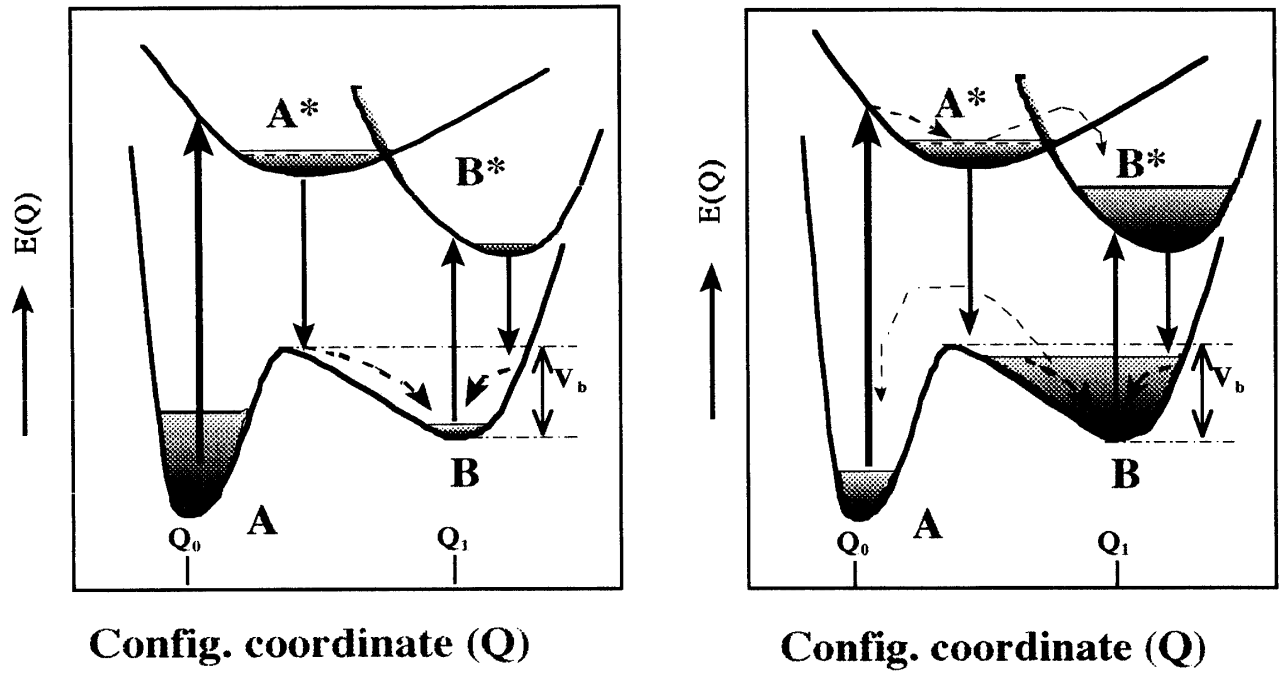

Fig.6 Configuration coordinate diagram. Left figure; before irradiation. Right figure; after irradiation . Arrows with the full lines; photoexcited transition or optical back reaction. Arrows with the dotted lines; thermal transitions.

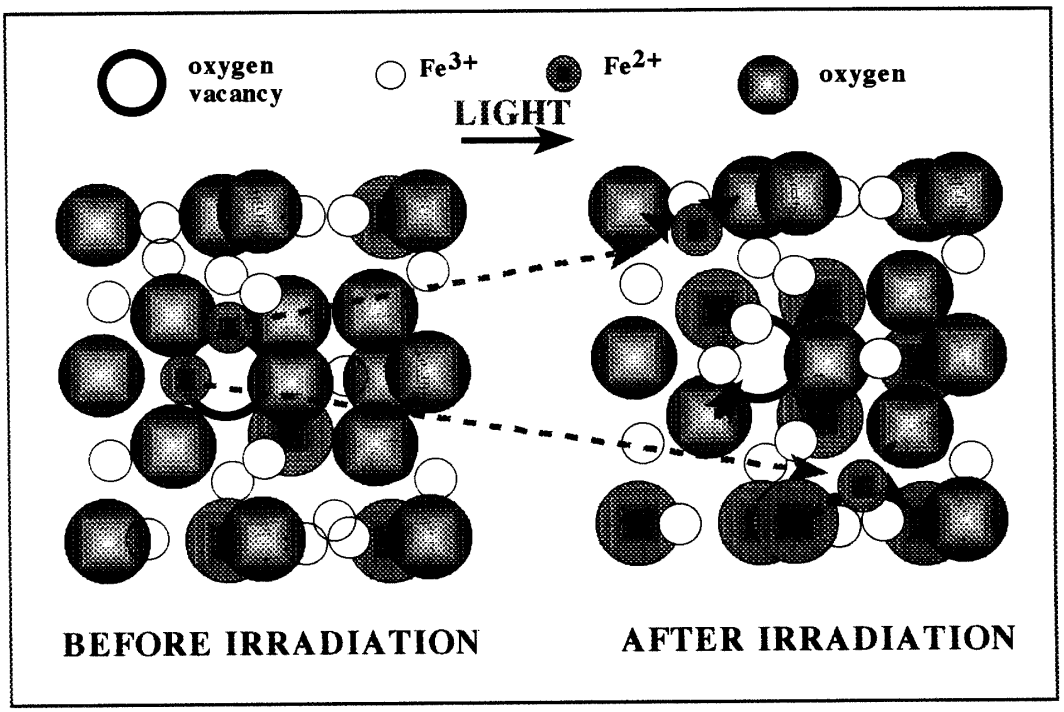

Fig.7. A schematic illustration of the photoinduced local distortion of lattice around oxygen vacancy.

V_b to the original state. The above discussion is based on the assumption that the strength of defect lattice coupling dependends critically on the localization of the electron or hole wavefunction at the defect. If during any transitionthis localization changes substantially (e.g. in transition from a localized state to a delocalized defect bound state or in charge transfer involving a localized defect), the lattice around the defects as this is assumed the case usually undergoes strong rearrangement (strong lattice relaxation). A configuration coordinate diagram is commonly used , which allows a good visualization of the most characteristic phenomena due to the strong defect lattice coupling. The total energy of the system E (electronic and elastic) is plotted versus the local lattice displacement represented by the so-called configuration coordinate Q(Fig.6). All defects in the materials can be divided into the two groups characterized either by weak or strong vibronic coupling. In other words a discontinuous change of the localization of the electronic wave function at the defect is observed. This problem has been discussed by several workers on the basis of different models of electron-phonon coupling[47]. Here, we repeat some key arguments given by Y.Toyozawa [47,48]. In the continum approximation the energy functional of a defect is given by

$$E(\lambda) = E_{kin}\ \lambda^2 - E_L\ \lambda - E_s\ \lambda^3 \quad , \qquad (1)$$

where λ is the degree of electron localization ($\lambda = a_0/a$: a_0-nerrest neighbour distance, a-electron effective radius), E_L is the sum of all long range forces (Coulomb) and Es- short range forces(localized potential+ acoustic phonon coiupling). It is obvious that depending on the relative strength of E_L and E_S we get either a delocalized ($\lambda << 1$ for $E_L >> E_S$) or a localized ($\lambda > 1$for $E_L < E_S$) state. The former is an effective Coulomb state, the latter the so-called extrinsic self-trapped state, the latter the so-called extrinsic self-trapped state. In the case of comparable binding energies of the two states they are separated by a barrier. Although the validity of the model is restricted to $\lambda < 1$, it can be simply extended to $\lambda > 1$ by an appropriate defect potential scaling ($m^* \rightarrow m_o$, $\varepsilon^* \rightarrow \varepsilon_\infty$) and proper inclusion of the local lattice distortion , e.g., the ground state of F-centers [47]. Electron localization means a strong lattice distortion around the defect as in the assumed case of oxygen vacancy. Such a localized state may exist only if the electronic binding energy is larger than the energy of the local lattice distortion. For most single impurities in covalent semiconductors this is not the case and the only stable state is the delocalized effective mass state with a relatively weak coupling to the lattice. In contrast, oxygen vacancy or defect aggregate provides a potential strong enough to bind electrons on a localized orbit with a resultant large local lattice distortion. So far the discussion is limitted to the situation assumed only for electrons or Fe^{2+} but may be extended for holes or Fe^{4+}. Figure 7 shows a schematic illustration of photoinduced local distortion of lattice around oxygen vacancy in a semi-real space. Accompanied with the experimental results so far mentioned above, a redistribution of the oscillator strength would occur (see appendix A) and display the various photoinduced magnetic or optical properties and behaviors. In order to obtain a direct support the problem, a detailed neutron diffraction experiment is in progress with a group of Professor T. Miyazaki of Tohoku University. However, a microscopic and quantitative description is quite complicated due to the interference of various interactions. The electrons and holes form a many particle system interacting with each other via Coulomb forces and magnetic interaction as well as

with lattice and impurities [see appendix B].

CONCLUSION

Photoinduced irreversible change in an optical absorption α is observed for a bulk single crystal and a thin film of yttrium-iron garnets (YIG) over the spectral range 600 or 700- 2000 nm and over the temperature range 80 -300 K. In some samples, coexistence of phtoinduced and dark $\Delta\alpha$-effect is found at low temperatures. And the sign of $\Delta\alpha$ is different from each other. This fact indicates this effect is not thermal effect but photoinduced effect. Experimental data are discussed within a simple phenomenological model, based on the configuration coordinate model.

Finally we thank for Professor K.Shinagawa of Toho University and Dr.T.Tamaki of Sci.Eng. Res. Center of NHK for fruitful discussions and warm encouragement.
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Appendix A:Oscillator Strength

Here, we summarize a fundamental concept of optical absorption phenomenon. The absorptioncoefficient α for a system of N atoms or centers per unit volume,is defined by the relation

$$dI = -\alpha I \, dx \qquad \text{(A-1)}$$

where I is the intensity of radiation , and dIi is the intensity cancelled from the beam in thickness dx. Thus α is the energy removed per unit time per unit volume from a beam of unit intensity. It clearly is equal to N times σ, the absorption cross section , and when written as a function of energy is given by

$$\alpha_{m->k}(E) = N \sigma_{m->k}$$

$$= \frac{4\pi^2 e^2}{3c} N r_{km}^{\ 2} \omega_{km} S_{mk}^{\ a}(E) \qquad \text{(A-2)}$$

The strength of atransition could be expressed in terms of its total absorption cross section or its decay time , but a more commonly used section but a more commonly used parameter is a dimensionless quantity, the oscillator strength. Classically, the polarizability of an oscillator of frequency ω is given by $e^2/m\omega^2$. Thus , the low-frequency polarizability of a classical atom in state m containing a series of oscillators of frequency ω_{km} with f_{mk} of each, can be

$$\alpha_{cl} = \sum_k e^2 f_{mk} / m\omega_{km}^{\ 2} \qquad \text{(A-3)}$$

Quantum-mechanically, the polarizability of an atom is easily found to be

$$\alpha_{qm} = 2\sum_k \frac{e^2 r_{km}^{\ 2}}{3(E_k - E_m)} \qquad \text{(A-4)}$$

by perturbation theory. By comparison, to make the two expressions agree, we define an effective number of oscillators of frequency ω km , i.e., the oscillator strength fmk , by the expression

$$f_{mk} = \frac{2m}{3h} \omega_{km} r_{km}^{\ 2} \qquad \text{(A-5)}$$

Strictly speaking, the oscillator strength is a tensor quantity, but it may be treated as a scalar in some simple cases. Its usefulnss in the study of optical properties is enhanced by theorems rgarding the total oscillator strength of an atom. For example, in the case of a one-electron atom(or center), the f-sum rule takes the particularly simple form

$$\sum_k f_{mk} = 1 \qquad \text{(A-6)}$$

Analogous relations exist in more complex systems. A few modifications need to be made in discussing the interaction of radiation with atoic systems embedded in a dielectric medium. The modifications tale a particularly simple form when the transitions of interest in the center occur at anenergy for which the medim is transparent and the centers are so far apart that interactions among them may be neglected. A few modifications must be made in discussing the interaction of radiation with atomic systems embedded in an insulating medium. The modifications take a particularly simple form when (a) the centers (in this case, oxygen vacancies) are not in resonance with the host medium, that is when the transition s of interest in the center occur at an energy for which the medium is transparent, and (b) the centers are so far diffuse that interactions among them may be neglected. In such case as in high pure YIG specimen, to which we shall largely restric ourselves, the presence of the other centers may be ignored, and each center may be treated as an isolated atom in a medium characterized by its real index of refraction, "effective mass" for charge carriers, and an effective mass. The wave functions and energy levels of the center are modified by the medium. In one extreme the influence of the neighboringatoms of the host crystal may be treated as a small perturbation, and in the opposite extrreme the wave functions of the medium itself are used to construct those for the center [49]. In order to express the macroscopic effects of the medium, it is well known that the transition amplitude is proportional to the field at the center (A_0). Thus the transition probability must be manltiplied by $(\varepsilon_{eff}/\varepsilon_0)^2$, where ε_{eff} is the magnitude of the field effective in inducing the transition, and ε_0 is the magnitude of the average field in the medium. In an ideal case, where the effective field ratio, that is $(\varepsilon_{eff}/\varepsilon_0)^2$ is known , the following relation may be considered as an expression for the experimental determination of the oscillator strength of the transition a->b. Alternatively, it may be used as a method for estimating the centers. The integrated absorption cross section may be expressed from a some tedeus calculation in the form as fllows,

$$\sum_{ab} = \left[\left(\varepsilon_{eff}/\varepsilon_0\right)^2\right]\left(\frac{1}{n}\right)\frac{2h\pi^2e^2}{cm^*} f_{ab} \quad . \text{(A-7)}$$

And then the integrated absorption coefficient may be obtained as follows,

$$\int \alpha_{ab}(E)dE = N\sum_{ab} \qquad \text{(A-8)}$$

Therefore , the following relation may be lead in a form,

$$Nf_{ab} = \frac{nm^*c}{2\pi^2e^2h}\left(\frac{\varepsilon_0}{\varepsilon_{eff}}\right)^2 \int \alpha_{ab}(E)dE \quad \text{(A-9)}$$

This equation is a generalized form of the well known Smakula's formula. For a center sufficiently well-localized state, Eq.(A-9) may be rewritten as

$$Nf_{ab} = 8.21\times10^{16}\, cm^{-3}\frac{n}{\left(n^2+2\right)^2}\int \alpha_{ab}(E)dE$$

(A-10)

where α is expressed in reciprocal centimeters and E in electron volts[50, 51].

Appendix B: Contribution of released electrons by photoexcitation

One of the authors (K.H) has been often asked where the released electrons may exist in a localized manner induced by photo-

excitation in YIG and what type of the behavior may be observable. Also based on the two center model, released electrons or Fe^{2+} ions may contribute to an anisotropy of these material in "normal environment" comparatively larger than near the centers. It is because the degeneracy of the energy level of Fe^{2+} is lifted near the center due to a low symmetry in the crystal. Attentively , in our context, we can explain the induced distortion by the released far away from oxygen vacancy in the real space. A question could be raised for a negatively charged irons or Fe^{2+} ions to be stable at such high temperature as 200 K since Fe^{3+} ion is effectively zero charged one in the crystal. In most cases placing an extra electron on a normal ion requires more energy due to the Coulomb repulsion between electrons. This added interaction energy is defined as U , which is often called the Hubbard correlation energy[52]. For almost all cases, U has a positive value. If an electron-phonon interaction ,however, stabilizes a surplus electron state more strongly than a single electron or normal state, then a quantity that governs the sequence of the energy levels is the difference between pure Coulomb electron-electron repulsion and the stabilization energy resulting from the electron-phonon interaction (e.g., due to the Jahn-Teller distortion) . Such an effective correlation energy U is effectively negative. We propose that this holds for the case of released electron besides nearest-neighbor of oxygen vacancy, although the negative-U phenomenon has been unambiguously proven.

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